Analysis of Carbon-14 Behavior for PWR Stations

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1. Introduction

Carbon-14 is a long-lived(T_H = 5730 yr) low-energy pure beta(E_{AVG} = 50 keV) emitter. A large amount of carbon-14 is released annually from nuclear power plants, and due to its long half-life and environmental mobility carbon-14 has long term radiological influence on environment.

So far, carbon-14 has been monitored and studied primarily at CANDU stations which discharge much more carbon-14 than BWR or PWR stations. However recently, annual individual doses to the general public from PWR normal operation are approaching the design objective values in reference [1] especially at multi-unit sites in Korea and carbon-14 is the main contributor to the normal operation doses.

Therefore, we are in urgent need of the carbon-14 monitoring and analysis programs for PWR stations in Korea. From last year, we have been studying this PWR carbon-14 issue and performing the analysis for PWR stations.

2. Analysis

In this section, the background data, model, and the application and verification of carbon-14 analysis for PWR stations are described in sequence.

2.1 Background Data

The background data for analysis consist of carbon-14 measurement data and plant operation data.

The measurement data presented here are from references [2] through [4].

The principal nuclear reactions forming carbon-14 for PWRs are neutron activation of oxygen-17 and nitrogen-14 in the reactor coolant, the former being the major sources.

- O-17(n,α)C-14
- N-14(n,p)C-14

The power-normalized production rate of carbon-14 for PWRs is about 10 Ci/GW(e)-yr.

Since the decay constant of carbon-14 is very small, most of carbon-14 produced in the reactor coolant system is removed either by purification ion-exchange resin or leakage, the latter corresponding to about 90 % for PWRs.

The specific activity of carbon-14 in the PWR reactor coolant system is about 1.0×10^4 μCi/g.

The chemical forms of carbon-14 in PWR reactor coolant, gaseous effluents, and ion-exchanger are predominantly organic, about 80 % being hydrocarbons (primarily methane) and others are inorganic, primarily carbon dioxide.

The following operation data for carbon-14 calculation are from reference [5] for YGN 5&6 units.

- reactor coolant mass, W = 1.97×10^8 g
- letdown flow rate, Q = 4723 g/sec
- letdown flow rate for boron control, B = 36 g/sec
- concentration of activation nuclides, atoms/g- H_2O
  C(O-17) = 1.3×10^{19}, C(N-14) = 7.3×10^{17}
- microscopic cross section, cm^2
  σ(O-17) = 1.5×10^{-25}, σ(N-14) = 1.16×10^{-24}
- in-core reactor coolant mass, m = 1.6×10^7 g
- neutron flux, $\phi = 6.6\times10^{13}$ n/cm^2-sec
- decay constant of C-14, $\lambda = 3.805\times10^{-12}$ /sec
- annual reactor operation time, T = 2.523×10^8 sec

2.2 Analysis Model

The differential equation for the quantity of carbon-14 ignoring removal by ion-exchange resin and leakage in the reactor coolant is as follows.

$$\frac{dN_R}{dt} = \sum C_i \sigma_i m \phi - \lambda N_R \quad (1)$$

where,
N_R : quantity of carbon-14

Solving equation (1) yields the following equation for carbon-14 annual production in the reactor coolant.

$$A_R(T) = \lambda N_R = \sum C_i \sigma_i m \phi [1 - \exp(-\lambda T)] \quad (2)$$

where,
A_R : annual production of carbon-14(Bq)

Since the decay constant of carbon-14 is extremely small, [1-exp(-λT)] ≈ λT and equation (2) can be reduced to the following equation.

$$A_R(T) = \lambda \sum C_i \sigma_i m \phi T \quad (3)$$

On the other hand, the differential equation for the quantity of carbon-14 considering the removal by ion-exchange resin and leakage in the reactor coolant is as follows.

$$\frac{dN_R}{dt} = \sum C_i \sigma_i m\phi - \lambda N_R - \frac{(Q-B)\eta}{W} N_R - \sum L_i N_R \quad (4)$$

where,
η : removal fraction of purification ion-exchanger
L : leak rate(/sec)

Here, the overall removal efficiency and leakage characteristics are assumed to be the same for organic and inorganic carbon-14, since the measurement data show that the chemical composition of carbon-14 is almost the same in reactor coolant, gaseous effluents, and ion-exchangers. Solving equation (4) yields the following equation for carbon-14 inventory in the reactor coolant.

$$I_R(t) = \lambda N_R(t) = \frac{\lambda \sum C_i \sigma_i m\phi}{\lambda + (Q-B)\eta / W + \sum L_i}\left[1 - \exp[-(\lambda + (Q-B)\eta / W + \sum L_i)t]\right] \quad (5)$$

where,
I_R : inventory of carbon-14(Bq)

The specific activity of carbon-14 in coolant is as follows.

$$S_R(t) = I_R(t)/W \quad (6)$$

where,
S_R : specific activity of carbon-14(Bq/g)

The inventory and specific activity of carbon-14 in the coolant system reach the values of the equilibrium state which are actually the same as those at the end of the operation time in a very short time after the beginning of power operation.

The build-up activity of carbon-14 on the ion-exchange resin ignoring the radioactive decay can be calculated as follows.

$$A_D(T) = \int_0^T S_R(t)(Q-B)\eta dt \approx S_R(T)(Q-B)\eta T \quad (7)$$

where,
A_D : build-up activity of carbon-14(Bq)

The integrated leakage amount of carbon-14 activity can be calculated as follows.

$$A_L(T) = \int_0^T S_R(t) W \sum L_i dt \approx S_R(T) W \sum L_i T \quad (8)$$

where,
A_L : integrated leakage of carbon-14(Bq)

The ion-exchanger removal and leakage amount of carbon-14 activity increase almost linearly with the operation time.

2.3 Application and Verification

First, we calculated carbon-14 annual production in YGN 5&6 reactor coolant system using equation (3) with the operation data given in section 2.1. The calculated value was 7.7 Ci/yr, which is in good agreement with the measurement value of about 10 Ci/GW(e)-yr.

Second, we calculated the specific activity of carbon-14 in the reactor coolant system using equation (5) and (6) with again the operation data in section 2.1. Here, the leak rate was assumed to be 9 times the removal rate by ion-exchange resin based on the measurement data in section 2.1. We calculated the values of the specific activity of carbon-14 in coolant varying the removal fractions(sensitivity analysis), and finally obtained the removal fraction corresponding to measurement value of 1.0×10^4 μCi/g, which is about 0.06. Here, the removal fraction for carbon-14 of PWR ion-exchange resin is the single-pass value averaged over the entire operation time, and is in agreement with the results of the scaled-down simulation experiment presented in reference [2].

3. Conclusion

We obtained the analysis model for carbon-14 behavior for PWR stations from the simple calculus and verified it by applying the model to a domestic PWR plant with its specific operation data and comparing the analysis results with the measurement data. The calculated values of carbon-14 annual production and specific activity from the analysis model were similar to those of the measurement data.

However, the application and verification of the analysis model here is based on the limited sources of carbon-14 measurement data available on PWRs, and therefore, more PWR data domestic and foreign, especially for removal mechanisms and overall efficiencies of purification ion-exchanger for organic and inorganic forms of carbon are necessary.

With more detailed and recent measurement data, we will be able to generalize the analysis model to be used for any PWR plants with their own specific operation data.

REFERENCES

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